

QUANTUM DYNAMICS IN MOLECULES AND NANOSTRUCTURES IN STRONG FIELDS

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Introduction. With the advance of time-dependent probes the investigation of the dynamical behavior of matter at the nanoscale has become a very important research direction. Laser-assisted steering of the electron motion in molecules and manipulation of their structure and composition has been dreamt about since the very emergence of coherent light sources. Realizing new possibilities in selective photochemistry of complex molecular systems, however, requires good understanding of underlying quantum dynamics. First-principles quantum-mechanical simulations can be of great help in assisting experiments, understanding the complex quantum dynamics, and interpreting the experimental data.

Computational method. We have developed a computational approach based on real-time real-space time-dependent density functional theory (TDDFT) [1]. In this approach, the Kohn-Sham orbitals are represented on numerical grids. The time evolution of the electrons is determined non-perturbatively and can handle strong external time-dependent fields. The motion of ions is treated classically using the Ehrenfest approach (Ehrenfest molecular dynamics). Complex absorbing potentials are implemented at the boundaries of the simulation volume in order to prevent unphysical reflections of the electron wave packets and allow longer time propagation.

Results and conclusions. We have used the developed computational approach to investigate several processes in molecules and nanostructures that involve strong field excitation, ionization, and fragmentation. Among these are the electron field emission from nanotubes [2], Coulomb explosion of small hydrocarbon molecules [3, 4], the interaction of energetic charged particles with graphene fragments [5], laser assisted desorption of atoms from surfaces [6], and others. The simulations are an indispensable tool to gain insight into the physical mechanisms behind highly nonequilibrium and nonlinear processes. The presented TDDFT framework is a low-cost and high-accuracy approach that can be used to simulate the electron and nuclear dynamics in strong external fields, to explain experimental results and to design new experiments.

References.

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